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STRATOSPHERIC OBSERVATIONS OF LONG-LIVED TRACE GASES AT MIDLATITUDES 1982–1985

(DATA REPORT)

by

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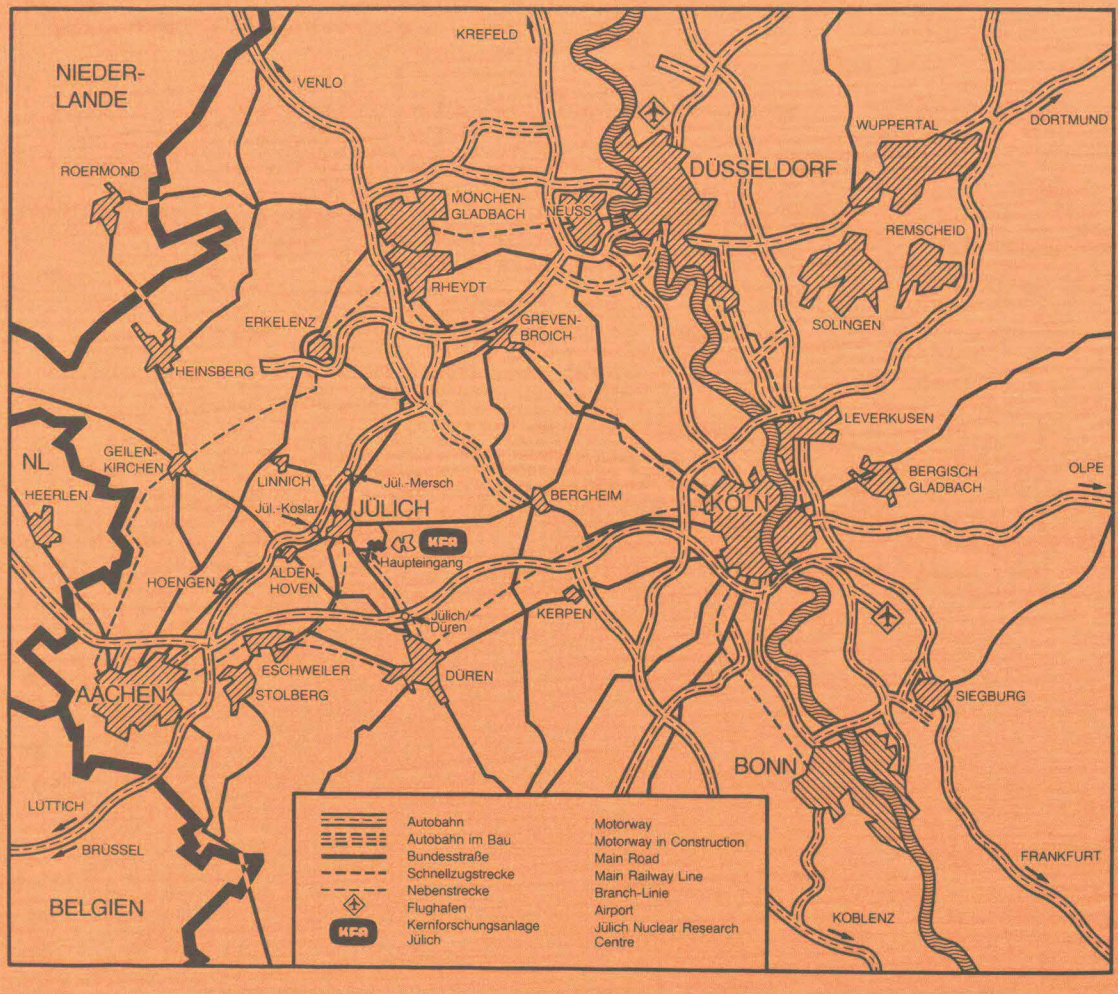

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1. Introduction

A field program to measure in situ the vertical distribution of long-lived trace gases in the stratosphere was begun in 1982. Large air samples were collected using a balloon-borne cryogenic whole-air sampler, that is cooled with liquid neon. Various gaschromatographic techniques were employed to determine the mixing ratios the following long-lived trace gases: CO_2, CH_4, CO, N_2O, $CFCl_3$, CF_2Cl_2, CH_3Cl, CH_3CCl_3, CCl_4, $C_2F_3Cl_3$. This report presents the data from seven balloon flights that were performed at midlatitudes over Southern France (44°C) in the time period from 1982-1985.

The results obtained during the individual flights are presented in tabulated form together with the relevant technical information for the respective flight. Plots of the combined data sets for the different species are also given.

This report is intended as a data report. It includes no interpretation or discussion and the measurements are not compared to any other data set. Part of the data has already been published. References are given in the sections describing the individual balloon flights.

2. Field experiments

The balloon flights have been performed at the two launching sites operated by the French Space Agency, C.N.E.S (Centre National D'Etudes Spatiales) at Gap (44°27'N; 06°02'E) and Aire s/l'Adour (43°42'N; 00°15'W) in Southern France. The program presented here is the continuation of a series of flights performed between 1977 and 1979 in cooperation with the MPI für Aeronomie, Lindau, FRG and the MPI für Chemie, Mainz, FRG (Fabian et al., 1971,1 1981; Volz et al., 1981). Most of the previous flights had been performed during the months of June. The recent program, therefore, was planned to allow for observations during different seasons of the year, in order to obtain additional information about the annual variability of the vertical distribution of the trace gases at midlatitudes.

The cryogenic whole air sampler is a newly designed improved version of the cryosamplers used by other groups (Lueb et l., 1975; Fabian et al., 1979). It allows to collect 15 samples of about 20 l (STP) during one balloon flight. A detailed description of this instrument is given elsewhere (Schmidt et al., 1983).

In general, samples were collected while the balloon was descending at a slow rate of 1 m/sec to 4 m/sec. The sample containers have seperate inlets that are located at the bottom of the cryosampler. They are evacuated in the laboratory prior to the flight and completely sealed until the inlet is opened by telecommand during the flight. Because the inlets are continously ventilated with clean ambient air during the descent, the risk of external contamination from the gondola is strongly reduced.

The new cryosampler differs from that one used during the previous program (Volz et al., 1981; Fabian et al., 1979) also with regard to another aspect. There is no "ozone filter" in the inlet tube. Such filters were suggested by Fabian et al. (1981), in order to avoid a possible alteration of the sample composition, when stratospheric air with an ozone content in the range of several ppmv is cryogenically collected into stainless steel container.

A laboratory simulation of stratospheric sampling showed that the concentrations of the trace gases investigated during this program are not affected by large amounts of ozone present (Knapska et al., 1985). It appears that the concentration of CCl_4 and CH_3CCl_3 slowly decays with time, however, even in the absence of ozone. Repeated analysis of original stratospheric samples (collected during flight BI (7) revealed that this systematic degradation amounts to less than 30 % if the analyses are completed within a time period of about four weeks after the sampling flight (Knapska, 1986). This requirement could be satisfied after each flight.

3. Laboratory analyses

After the flight the sampler is transported back to the laboratory. After warm-up the sample pressure in the cryopumps increases rapidly to about 50 bar, depending on the collected sample volume. At such high pressures molecular diffusion is rather slow. Most of the species to be measured, especially the halogenated hydrocarbons have rather high boiling temperatures. Therefore it takes almost one week before the sample is homogenized again. The measurements of the various trace gases were performed by using four different gaschromatographic techniques.

a) CH_4, CO and CO_2 are analysed in one chromatograph with a dual column system. CH_4 and CO as well as CO_2 are separated on molecular sieve (5 Å) and porous silica, respectively. Both columns are kept at 380 K. A sample volume of 2-5 cm^3 (CO and CH_4) or 1 cm^3 (CO_2) is needed for one analysis run. After separation both CO and CO_2 are catalytically converted to CH_4, employing a hot Ni-catalyst (620 K) in the presence of H_2, prior to detection with an flame-ionization detector.

b) N_2O, $CFCl_3$, and CF_2Cl_2 are separated on Porasil C. The column is temperature programmed between 240 K and 370 K. High purity dry nitrogen is used as carier gas. The compounds are detected with an Electron Capture Detector (ECD) kept at 580 K. The sample volume is 2-5 cm^3 per chromatographic run.

c) The halogenated hydrocarbons CCl_4, CH_3CCl_3, CH_3Cl, and $C_2F_3Cl_3$ are first preconcentrated at 70 K from sample volumes of 1-2 liters on a short column filled with glass beads. Thereafter they are transferred onto the main column and seperated on Durapak Porasil C/n-octane, temperature programmed between 170 K and 370 K, using high purity dry nitrogen as carrier gas, doped with $\sim$ 1.5 % of pure oxygen. An Electron Capture Detector (ECD), kept at 570 K is used for detection.

4. Calibration

For calibration, the samples were compared to laboratory standards. These are large tropospheric air samples, that were cryogenically collected at a remote site. They were diluted with purified synthetic air to adjust their trace gas concentrations to mixing ratios that correspond to values observed in the lower strosphere. These laboratory standards were in turn compared against absolutely calibrated reference standards.

For CO, CH_4, N_2O and for the chlorofluorocarbons $CFCl_3$ and CF_2Cl_2 the laboratory standards in use during this series of observations can be traced backwords to the reference standards used ten years ago. Their absolute calibration has been repeatedly checked by comparison with reference standards prepared by three step static dilution of these compounds in "zero" synthetic air. The accuracy for these standards is $\pm$5 %, except for $CFCl_3$, for which it is estimated to $\pm$10 % (Volz et al., 1981). The CO_2 measurements also rely on the same laboratory standard, that has been in use since 1976. This standard

has been repeatedly compared to the Scripps Manometric Scale at the Institut für Umweltphysik, Heidelberg, F.R.G. during recent years. It was found to be about 2 ppm lower than the Manometric Scale.

During the first part of this programm, an absolute calibration for the halocarbons CH_3Cl, CCl_4, CH_3CCl_3 and $C_2F_3Cl_3$ was not available. Therefore our laboratory standards were compared to the "Harwell Standard" that was provided by S.A. Penkett. After the analyses at Jülich most of the stratospheric samples from flights BI/3 were also analyzed at Harwell using the gaschromatographs/mass spectrometer combination (Knapska et al., 1985; Hough et al., 1986). It should be noted that the stratospheric measurements of these four halocarbons made until 1984 all rely on the AERE-Harwell absolute calibration (Borchers et al., 1983; Penkett et al., 1980; Schmidt et al., 1980, 1984, 1985).

Since 1984 a new laboratory standard has been used, that had been very carefully compared to the Atmospheric Lifetime Experiment (ALE) calibration standards (Rasmussen, 1984). The absolute calibration of the halocarbon species is quite difficult to establish and has only been relatively poorly defined yet (Rasmussen and Lovelock, 1983). However, it appears that the relative calibration of the ALE standard can be made at a rather high precision. We report our measurements relative to this ALE type standard. The absolute values are subject to a possible future change of the calibration factors for the ALE standards (Rasmussen and Lovelock, 1983).

For the various halocarbons the uncertainty in the absolute calibration is estimated to less than $\pm$ 10 %, except for CCl_4, for which an accuracy of $\pm$20 % is estimated (Rasmussen and Lovelock, 1983).

In order to sparingly use the volume of the collected air samples, the analytical precision is determined from the series of runs of the laboratory standard rather than from repeated analysis of original air samples. The precision ($\pm 1\sigma$) is $\pm$0.3 % for CO_2, $\pm$1 % for CH_4, and less than $\pm$3 % for N_2O, CO, $CFCl_3$, and CF_2Cl_2, respectively. For the other halocarbons this relative errors amounts to about $\pm$10 %. The actual errors listed in the data tables represent the precision or the lower limit of detection, whichever is larger. The detection limit is calculated from the detector signals obtained during a standard run and assuming a signal to noise ratio S/N = 2.

5. Results

The results obtained during seven balloon flights are presented in Tables 1 through 7. Each of the tables consists of three parts.

Part A: General information and technical flight data.
Part B: Measurements of CO, CO_2, CH_4, N_2O, $CFCl_3$, and CF_2Cl_2.
Part C: Measurements of CH_3Cl, CH_3CCl_3, CCl_4, and $C_2F_3Cl_3$.

Ambient temperature was measured by means of a VIZ Accu-Lok temperature sensor. This thermistor type sensor was mounted on the end of a beam (1 m long) extending from the gondola frame to ensure proper ventilation. Ozone was monitored with a Mast Model 730-8 Ozonesonde. It must be noted that the ozone measurements were not made according to the standards of the ozonesonde network, because the ascent rate of large scientific balloons is slower ($\sim$3 m/s) than that of a standard radiosonde ($\sim$5 m/s).

Figures 1 through 7 display the in situ observations of the various species in graphical form. Note that the altitude scale is not the true geometric height. The primary measurement of altitude is the ambient pressure which was converted to the geometric altitude using the U.S. Standard Atmosphere (1976). The ambient pressure was measured during the flights by the 'CNES' balloon launch facility employing two Penny and Giles pressure transducers (type TP 71 A/200 with ranges from 1634 to 10 mb and from 69 to 0 mb. The error of these transducers is 1 % of reading and 0.7 mb respectively (A. Valette, 1982, priv. communication). This corresponds to a maximum error in the vertical scale of about 600 m at 33 km altitude (7.7 mb). At altitudes below the 10 mb level the sampling intervals are smaller than 1 km. Even at higher altitudes they do not exceed 2.5 km. The tables give the actual upper and lower boundaries for the individual samples. In the figures, however, the symbols are plotted at sampling altitudes, that correspond to the geometric average of the pressure within the sampling intervals. In order to maintain clarity neither error bars nor the sampling intervals are plotted.

6. Acknowledgements

The balloon flights were funded by the Bundesministerium für Forschung und Technologie (BMFT) through grants FKW 25 and KBF 66. We are indebted to the balloon launching team of the French CNES for the excellent performance of the balloon launches.

Table 1 A:

Flight No.:	BI/1
Date:	10. June 1982
Size of balloon:	100.000 m^3 (Zodiak)
Location:	Gap-Tallard
Launch time (GMT):	04:57 a.m.
Duration of flight:	11 h 21 min
Tropopause pressure:	175 mb
altitude:	12.7 km
temperature:	-65°C

The results are listed in table 1 B.

References: Schmidt et al. (1984)

Comments: This was a technological flight to test the performance of the newly developed cryosampler. Various in flight tests were performed to check for sample integrity during sampling. Therefore the number of samples was limited.

Table 1 B:

Flight-No.: BI/1 Date: 10. June 1982

Sample No.	Altitude [mb]	Altitude [km]*	T [°C]	O_3 [ppmv]	CO [ppbv]	CO_2 [ppmv]	CH_4 [ppmv]	N_2O [ppbv]	$CFCL_3$ [pptv]	CF_2Cl_2 [pptv]
1	20.4 - 22.1	25.9 - 26.5	-	-	17 ± 5	331 ± 1	0.96 ± 0.01	106 ± 5	2.4 ± 0.1	89 ± 2
2	40.5 - 43.6	21.5 - 22.0	-	-	20 ± 5	329 ± 1	1.10 ± 0.01	195 ± 6	23.0 ± 0.7	140 ± 3
3	58.5 - 62.4	19.2 - 19.6	-	-	18 ± 5	333 ± 1	1.21 ± 0.02	230 ± 7	57.0 ± 1.7	179 ± 4

* US-Standard Atmosphere 1976

Table 2 A:

Flight No.:	BI/2
Date:	21. October 1982
Size of balloon:	100.000 m^3 (Zodiak)
Location:	Aire s/l'Adour
Launch time (GMT):	07:51 a.m.
Duration of flight:	6 h 52 min
Tropopause pressure:	125 mb
altitude:	14.8 km
temperature:	-63.9°C

The results are listed in table 2 B, 2 C.

References: Schmidt et al. (1983; 1984; 1985)
Hough et al. (1986)
Knapska et al. (1985)
Borchers et al. (1985)

Comments:

Table 2 B:

Flight-No.: BI/2 Date: 21. October 1982

Sample No.	Altitude [mb]	Altitude [km]*	T [°C]	O_3 [ppmv]	CO [ppbv]	CO_2 [ppmv]	CH_4 [ppmv]	N_2O [ppbv]	$CFCL_3$ [pptv]	CF_2Cl_2 [pptv]
1	7.2 - 7.3	33.2 - 33.3	-39.0	-	11 ± 5	332 ± 1	0.51 ± 0.01	21 ± 5	<0.2	4.7 ± 0.5
2	7.9 - 8.5	32.3 - 32.8	-42.3	-	14 ± 5	331 ± 1	0.53 ± 0.01	23 ± 5	<0.2	5.6 ± 0.5
3	10.2 - 10.6	30.8 - 31.1	-47.3	-	9 ± 5	330 ± 1	0.62 ± 0.01	43 ± 5	<0.2	15.4 ± 0.5
4	18.3 - 18.4	27.1 - 27.2	-55.0	-	26 ± 5	332 ± 1	0.93 ± 0.01	117 ± 5	1.0 ± 0.2	74.0 ± 1.5
5	22.8 - 23.1	25.6 - 25.7	-55.2	-	54 ± 5	330 ± 1	0.98 ± 0.01	149 ± 5	4.9 ± 0.2	100.0 ± 2.0
6	27.5 - 27.7	24.4 - 24.5	-55.3	-	67 ± 5	332 ± 1	0.98 ± 0.01	157 ± 5	10.7 ± 0.3	111.0 ± 2.5
7	33.9 - 35.2	22.9 - 23.1	-59.3	2.75	9 ± 5	331 ± 1	1.00 ± 0.01	169 ± 5	21.6 ± 0.6	124.0 ± 2.5
8	43.9 - 45.7	21.2 - 21.5	-59.9	2.08	7 ± 5	332 ± 1	1.08 ± 0.01	191 ± 6	40.0 ± 1.2	149.0 ± 3.0
9	70.0	18.5	-63.3	0.84	5 ± 5	337 ± 1	1.47 ± 0.01	285 ± 8	125.0 ± 3.8	263.0 ± 5.3
10	89.0	17.0	-62.3	0.41	14 ± 5	337 ± 1	1.48 ± 0.02	297 ± 9	143.0 ± 5.0	280.0 ± 5.6
11	110.0 - 111.0	15.6	-63.3	0.21	7 ± 5	334 ± 1	1.61 ± 0.02	307 ± 9	165.0 ± 5.0	305.0 ± 6.1

* US-Standard Atmosphere 1976

Table 2 C:

Flight-No. BI/2 Date: 21. October 1982

Sample No.	Altitude [mb]	Altitude [km]*	CH_3Cl [pptv]	CH_3CCl_3 [pptv]	CCl_4 [pptv]	$C_2F_3Cl_3$ [pptv]
1	7.2 - 7.3	33.2 - 33.3	26 ± 5	<0.5	<0.05	<0.2
2	7.9 - 8.5	32.3 - 32.8	23 ± 5	<0.5	≦0.05	0.2 ± 0.2
3	10.2 - 10.6	30.8 - 31.1	20 ± 5	<0.5	0.13 ± 0.05	0.7 ± 0.2
4	18.3 - 18.4	27.1 - 27.2	76 ± 8	3.0 ± 0.5	0.22 ± 0.05	5.3 ± 0.5
5	22.8 - 23.1	25.6 - 25.7	103 ± 10	4.5 ± 0.5	0.74 ± 0.07	7.6 ± 0.8
6	27.5 - 27.7	24.4 - 24.5	135 ± 15	3.7 ± 0.5	2.40 ± 0.25	8.7 ± 0.9
7	33.9 - 35.2	22.9 - 23.1	115 ± 12	7.4 ± 0.8	4.6 ± 0.46	11.0 ± 1.1
8	43.9 - 45.7	21.2 - 21.5	135 ± 15	15.0 ± 1.8	10.0 ± 1.0	13.0 ± 1.3
9	70.0	18.5	340 ± 35	52.0 ± 6.0	43.0 ± 4.3	30.0 ± 3.0
10	89.0	17.0	445 ± 50	56.0 ± 6.5	37.0 ± 3.7	31.0 ± 3.1
11	110.0 - 111.0	15.6	510 ± 55	68.0 ± 7.5	72.0 ± 5.9	34.0 ± 3.4

* US-Standard Atmosphere 1976

Table 3 A:

Flight No.:	BI/3
Date:	10. September 1983
Size of balloon:	100.000 m^3 (Zodiak)
Location:	Aire s/l'Adour
Launch time (GMT):	04:15 a.m.
Duration of flight:	6 h 53 min
Tropopause pressure:	174 mb
altitude:	12.7 km
temperature:	-59.4°C

The results are listed in table 3 B, 3 C.

References:

Schmidt et al. (1984; 1985; 1986)
Knapska et al. (1985)
Hough et al. (1986)
Borchers et al. (1986)

Comments:

This flight was part of the MAP-Globus Campaign 1983. The instrument was launchED on gondola G9 to perform a field intercomparison of the cryo-samplers in use at the MPI for Aeronomie, Lindau and at the KFA.

Table 3 B:

Flight-No.: BI/3 Date: 10. September 1983

Sample No.	Altitude [mb]	Altitude [km]*	T [°C]	O_3 [ppmv]	CO [ppbv]	CO_2 [ppmv]	CH_4 [ppmv]	N_2O [ppbv]	$CFCl_3$ [pptv]	CF_2Cl_2 [pptv]
1	5.9 - 6.3	34.3 - 34.8	-40.7	-	5 ± 5	331 ± 1	0.82 ± 0.01	51 ± 5	<0.2	16 ± 1
2	7.1 - 8.6	32.2 - 33.5	-42.5	-	11 ± 5	329 ± 1	0.75 ± 0.01	56 ± 5	<0.2	20 ± 1
3	8.7 - 10.2	31.0 - 32.1	-42.5	-	54 ± 5	329 ± 1	0.83 ± 0.01	59 ± 5	<0.2	22 ± 1
4	10.2 - 11.7	30.1 - 31.0	-45.5	-	<5	331 ± 1	0.79 ± 0.01	68 ± 5	<0.2	26 ± 1
5	13.2 - 14.8	28.6 - 29.3	-47.0	-	9 ± 5	331 ± 1	0.89 ± 0.01	93 ± 5	<0.2	47 ± 1
6	16.4 - 16.9	27.7 - 27.9	-47.5	-	13 ± 5	330 ± 1	0.94 ± 0.01	108 ± 5	1.3 ± 0.2	58 ± 1
7	20.2 - 21.4	26.1 - 26.5	-49.5	-	26 ± 5	329 ± 1	0.90 ± 0.01	106 ± 5	2.1 ± 0.2	58 ± 1
8	24.5 - 25.1	25.1 - 25.3	-50.1	∿3.0	16 ± 5	330 ± 1	0.93 ± 0.01	113 ± 5	3.7 ± 0.2	65 ± 2
9	30.4 - 32.8	23.4 - 23.8	-54.8	3.31	6 ± 5	330 ± 1	0.91 ± 0.01	119 ± 5	8.3 ± 0.3	75 ± 2
10	36.9 - 39.0	22.2 - 22.6	-54.7	3.21	5 ± 5	330 ± 1	1.08 ± 0.01	175 ± 5	37.0 ± 0.7	131 ± 3
11	46.2 - 48.5	20.8 - 21.1	-56.0	2.45	5 ± 5	331 ± 1	1.23 ± 0.01	217 ± 6	74.0 ± 1.5	182 ± 3
12	58.1 - 59.7	19.5 - 19.7	-57.0	1.46	15 ± 5	333 ± 1	1.38 ± 0.02	256 ± 8	109.0 ± 2.0	228 ± 3
13	68.0	18.6	-57.0	1.08	19 ± 5	335 ± 1	1.44 ± 0.02	270 ± 8	127.0 ± 2.5	249 ± 4
14	83.0 - 86.0	17.1 - 17.3	-56.5	0.76	5 ± 5	332 ± 1	1.46 ± 0.02	290 ± 9	145.0 ± 2.9	273 ± 4
15	110.0 - 113.0	15.4 - 15.5	-58.5	0.38	21 ± 5	334 ± 1	1.54 ± 0.02	316 ± 10	170.0 ± 3.5	297 ± 6

* US-Standard Atmosphere 1976

Table 3 C:

Flight-No. BI/3 Date: 10. September 1983

Sample No.	Altitude [mb]	Altitude [km]*	CH_3Cl [pptv]	CH_3CCl_3 [pptv]	CCl_4 [pptv]	$C_2F_3Cl_3$ [pptv]
1	5.9 - 6.3	34.3 - 34.8	22 ± 5	<1.0	<0.04	0.7 ± 0.2
2	7.1 - 8.6	32.2 - 33.5	6 ± 5	<1.0	<0.04	0.8 ± 0.2
3	8.7 - 10.2	31.0 - 32.1	13 ± 5	<1.0	<0.04	0.9 ± 0.2
4	10.2 - 11.7	30.1 - 31.0	8 ± 5	<1.0	<0.04	1.3 ± 0.2
5	13.2 - 14.8	28.6 - 29.3	21 ± 5	<1.0	<0.04	2.6 ± 0.2
6	16.4 - 16.9	27.7 - 27.9	20 ± 5	<1.0	-	4.2 ± 0.2
7	20.2 - 21.4	26.1 - 26.5	16 ± 5	-	0.05± 0.04	2.6 ± 0.2
8	24.5 - 25.1	25.1 - 25.3	45 ± 5	1.2 ± 1.0	0.6 ± 0.15	4.6 ± 0.2
9	30.4 - 32.8	23.4 - 23.8	33 ± 5	1.2 ± 1.0	1.1 ± 0.25	5.2 ± 0.3
10	36.9 - 39.0	22.2 - 22.6	76 ± 9	15.0 ± 2.5	12.0 ± 2.5	10.0 ± 0.5
11	46.2 - 48.5	20.8 - 21.1	140 ± 16	34.0 ± 5.1	22.0 ± 4.8	14.0 ± 0.7
12	58.1 - 59.7	19.5 - 19.7	245 ± 30	42.0 ± 6.3	32.0 ± 7.0	20.0 ± 1.0
13	68.0	18.6	270 ± 32	66.0 ± 10.0	43.0 ± 9.5	20.0 ± 1.0
14	83.0 - 86.0	17.1 - 17.3	260 ± 31	87.0 ± 13.0	61.0 ± 13.4	26.0 ± 1.3
15	110.0 - 113.0	15.4 - 15.5	325 ± 39	99.0 ± 15.0	59.0 ± 13.0	28.0 ± 1.4

* US-Standard Atmosphere 1976

Table 4 A:

Flight No.:	BI/4
Date:	13. April 1984
Size of balloon:	100.000 m^3 (Zodiak)
Location:	Aire s/l'Adour
Launch time (GMT):	06:19 a.m.
Duration of flight:	6 h 51 min
Tropopause pressure:	186 mb
altitude:	12.3 km
temperature:	-63.6°C

The results are listed in table 4 B.

Comments: Data on halocarbons not available.

Table 4 B:

Flight-No. BI/4 Date: 13. April 1984

Sample No.	Altitude [mb]	Altitude [km]*	T [°C]	O_3 [ppmv]	CO [ppbv]	CO_2 [ppmv]	CO_4 [ppmv]	N_2O [ppbv]	$CFCl_3$ [pptv]	$CFCl_2$ [pptv]
1	7.9 - 9.2	31.8 - 32.8	-39.2	-	18 ± 5	332 ± 1	0.78 ± 0.01	58 ± 5	<0.2	30 ± 1
2	9.4 - 10.9	30.6 - 31.6	-43.7	-	17 ± 5	334 ± 1	0.71 ± 0.01	56 ± 5	<0.2	33 ± 1
3	11.4 - 13.2	29.3 - 30.3	-47.8	-	15 ± 5	330 ± 1	0.91 ± 0.01	102 ± 5	1.0 ± 0.2	65 ± 2
4	15.9 - 17.6	27.4 - 28.1	-50.1	-	<5	334 ± 1	1.05 ± 0.01	118 ± 5	3.0 ± 0.2	84 ± 2
5	23.0 - 24.1	25.4 - 25.7	-53.5	3.9	17 ± 5	331 ± 1	1.06 ± 0.01	134 ± 5	6.5 ± 0.2	90 ± 2
6	28.1 - 29.5	24.0 - 24.3	-56.1	3.7	11 ± 5	330 ± 1	0.99 ± 0.01	122 ± 5	11.9 ± 0.4	90 ± 2

* US-Standard Atmosphere 1976

Table 5 A:

Flight No.:	BI/5
Date:	27. September 1984
Size of balloon:	350.000 m^3 (Zodiak)
Location:	Aire s/l'Adour
Launch time (GMT):	06:43 a.m.
Duration of flight:	5 h 47 min
Tropopause pressure:	191 mb
altitude:	12.1 km
temperature:	-62.9°C

The results are listed in table 5 B, 5 C.

Table 5 B:

Flight-No. BI/5 Date: 27. September 1984

Sample No.	Altitude [mb]	Altitude [km]*	T [°C]	O_3 [ppmv]	CO [ppbv]	CO_2 [ppmv]	CH_4 [ppmv]	N_2O [ppbv]	$CFCl_3$ [pptv]	CF_2Cl_2 [pptv]
1§	20.4 - 23.6	25.5 - 26.5	-50.0	-	9 ± 5	332 ± 1	1.01 ± 0.01	127 ± 5	11.5 ± 0.4	97 ± 2
2	3.3 - 4.0	37.4 - 39.0	-26.1	-	42 ± 5	331 ± 1	0.71 ± 0.01	-	-	-
3					51 ± 5	332 ± 1	0.69 ± 0.01	-	-	-
4	4.0 - 5.6	35.2 - 37.4	-33.1	-	15 ± 5	332 ± 1	0.65 ± 0.01	-	-	-
5					18 ± 5	333 ± 1	0.67 ± 0.01	-	-	-
6	5.6 - 7.0	33.6 - 35.2	-39.0	-	14 ± 5	333 ± 1	0.75 ± 0.01	-	-	-
7	7.1 - 10.0	31.2 - 33.5	-41.3	-	29 ± 5	333 ± 1	0.78 ± 0.01	63 ± 5	<0.2	30 ± 1
8					70 ± 5	332 ± 1	0.80 ± 0.01	-	-	-

* US-Standard Atmosphere 1976

§ collected during ascent

Table 5 C:

Flight-No. BI/5 Date: 27. September 1984

Sample No.	Altitude [mb]	Altitude [km]*	CH_3Cl [pptv]	CH_3CCl_3 [pptv]	CCl_4 [pptv]	$C_2F_3Cl_3$ [pptv]
1§	20.4 - 23.6	25.5 - 26.5	38 ± 10	<0.5	<0.2	3.2 ± 0.30
2	3.3 - 4.0	37.4 - 39.0	-	-	-	-
3			-	-	-	-
4	4.0 - 5.6	35.2 - 37.4	17 ± 5	<0.5	<0.2	0.21 ± 0.02
5			10 ± 5	<0.5	<0.2	0.19 ± 0.02
6	5.6 - 7.0	33.6 - 35.2	21 ± 5	<0.5	<0.2	0.37 ± 0.04
7	7.1 - 10.0	31.2 - 33.5	31 ± 8	<0.5	<0.2	0.79 ± 0.08
8			38 ± 10	<0.5	<0.2	0.80 ± 0.08

* US-Standard Atmosphere 1976

§ collected during ascent

Table 6 A:

Flight No.:	BI/7
Date:	31. March 1985
Size of balloon:	100.000 m^3 (Winzen)
Location:	Aire s/l'Adour
Launch time (GMT):	07:28 a.m.
Duration of flight:	5 h 52 min
Tropopause pressure:	243 mb
altitude:	10.5 km
temperature:	-63.7°C

The results are listed in table 6 B, 6 C.

Table 6 B:

Flight-No. BI/7c Date: 31. March 1985

Sample No.	Altitude [mb]	Altitude [km]*	T [°C]	O_3 [ppmv]	CO [ppbv]	CO_2 [ppmv]	CH_4 [ppmv]	N_2O [ppbv]	$CFCl_3$ [pptv]	CF_2Cl_2 [pptv]
1	8.0 - 8.6	32.2 - 32.7	-34.5	5.4	<5	333 ± 1	0.66 ± 0.01	41 ± 5	<0.1	16 ± 1
2	10.0 - 11.5	30.3 - 31.2	-36.5	5.3	29 ± 5	334 ± 1	0.66 ± 0.01	41 ± 5	2.2 ± 0.1	24 ± 1
3	12.0 - 13.9	29.0 - 30.0	-41.2	6.0	52 ± 5	334 ± 1	0.71 ± 0.01	49 ± 5	<0.1	19 ± 1
4	15.0 - 17.0	27.7 - 28.5	-47.8	4.3	8 ± 5	331 ± 1	0.63 ± 0.01	34 ± 5	1.4 ± 0.1	17 ± 1
5	17.1 - 19.5	26.8 - 27.6	-50.9	4.5	11 ± 5	331 ± 1	0.70 ± 0.01	44 ± 5	0.8 ± 0.1	21 ± 1
6	21.0 - 23.0	25.7 - 26.3	-52.9	4.5	14 ± 5	334 ± 1	0.90 ± 0.01	94 ± 5	1.1 ± 0.1	56 ± 1
7	28.0 - 29.5	24.0 - 24.4	-58.2	3.5	<5	334 ± 1	1.05 ± 0.01	147 ± 5	17.0 ± 0.5	124 ± 3
8	35.0 - 36.0	22.8 - 22.9	-60.7	3.2	268 ± 5	335 ± 1	1.20 ± 0.01	181 ± 5	34.0 ± 1.0	142 ± 3
9	44.7 - 49.1	20.8 - 21.4	-62.5	2.3	20 ± 5	337 ± 1	1.31 ± 0.02	231 ± 7	73.0 ± 2.2	206 ± 6
10	55.7 - 61.0	19.4 - 20.0	-63.4	1.6	13 ± 5	336 ± 1	1.36 ± 0.02	254 ± 8	92.0 ± 3.0	225 ± 6
11	70 - 74	18.1 - 18.5	-60.8	1.1	26 ± 5	335 ± 1	1.43 ± 0.02	268 ± 8	116.0 ± 3.5	262 ± 8
12	82 - 86	17.2 - 17.5	-59.6	1.2	17 ± 5	335 ± 1	1.44 ± 0.02	261 ± 8	113.0 ± 3.5	254 ± 8
13	102 - 106	15.9 - 16.1	-59.9	0.8	33 ± 5	330 ± 1	1.46 ± 0.02	275 ± 8	121.0 ± 3.6	262 ± 8
14	119 - 123	14.9 - 15.1	-58.5	0.6	11 ± 5	334 ± 1	1.47 ± 0.02	268 ± 8	119.0 ± 3.5	262 ± 8
15	146 - 148	13.7 - 13.8	-57.1	0.3	45 ± 5	338 ± 1	1.60 ± 0.02	307 ± 9	158.0 ± 4.5	313 ± 9

* US-Standard Atmosphere 1976

Table 6 C:

Flight-No. BI/7c Date: 31. March 1985

Sample No.	Altitude [mb]	Altitude [km]*	CH_3Cl [pptv]	CH_3CCl_3 [pptv]	CCl_4 [pptv]	$C_2F_3Cl_3$ [pptv]
1	8.0 - 8.6	32.2 - 32.7	22 ± 4.5	<0.2	<0.02	0.4 ± 0.02
2	10.0 - 11.5	30.3 - 31.2	21 ± 4.0	<0.1	0.04 ± 0.01	0.4 ± 0.02
3	12.0 - 13.9	29.0 - 30.0	16 ± 3.0	<0.1	0.01 ± 0.01	0.5 ± 0.02
4	15.0 - 17.0	27.7 - 28.5	10 ± 2.0	<0.1	<0.01	0.3 ± 0.02
5	17.1 - 19.5	26.8 - 27.6	15 ± 3.0	<0.2	<0.02	0.6 ± 0.06
6	21.0 - 23.0	25.7 - 26.3	37 ± 7.0	<0.3	0.2 ± 0.03	2.3 ± 0.23
7	28.0 - 29.5	24.0 - 24.4	112 ± 22.0	7.4 ± 0.1	6.9 ± 1.1	6.1 ± 0.61
8	35.0 - 36.0	22.8 - 22.9	74 ± 15.0	15.0 ± 1.5	12.0 ± 2.0	7.4 ± 0.74
9	44.7 - 49.1	20.8 - 21.4	152 ± 30.0	40.0 ± 3.6	36.0 ± 6.0	11.0 ± 1.10
10	55.7 - 61.0	19.4 - 20.0	163 ± 33.0	59.0 ± 5.5	49.0 ± 8.0	14.0 ± 1.40
11	70 - 74	18.1 - 18.5	219 ± 44.0	71.0 ± 6.5	63.0 ± 10.0	14.0 ± 1.40
12	82 - 86	17.2 - 17.5	320 ± 65.0	74.0 ± 6.5	67.0 ± 11.0	16.0 ± 1.60
13	102 - 106	15.9 - 16.1	343 ± 68.0	81.0 ± 7.0	77.0 ± 13.0	17.0 ± 1.70
14	119 - 123	14.9 - 15.1	416 ± 83.0	76.0 ± 7.0	67.0 ± 11.0	17.0 ± 1.70
15	146 - 148	13.7 - 13.8	410 ± 82.0	96.0 ± 9.0	83.0 ± 14.0	19.0 ± 1.90

* US-Standard Atmosphere 1976

Table 7 A:

Flight No.:	BI/8
Date:	21. October 1985
Size of balloon:	100.000 m³ (Zodiak)
Location:	Aire s/l'Adour
Launch time (GMT):	08:47 a.m.
Duration of flight:	7 h 43 min
Tropopause pressure:	199 mb
altitude:	11.8 km
temperature:	-62.2°C

The results are listed in table 7 B, 7 C.

Table 7 B:

Flight-No. BI/8 Date: 21. October 1985

Sample No.	Altitude [mb]	Altitude [km]*	T [°C]	O_3 [ppmv]	CO [ppbv]	CO_2 [ppmv]	CH_4 [ppmv]	N_2O [ppbv]	$CFCl_3$ [pptv]	CF_2Cl_2 [pptv]
1§	15.0 - 21.9	26.0 - 28.5	-51.7	-	7 ± 5	333 ± 1	0.82 ± 0.01	87 ± 5	0.70 ± 0.2	55 ± 1
2	7.3 - 8.1	32.6 - 33.6	-43.6	-	<5	333 ± 1	0.48 ± 0.01	18 ± 5	0.02 ± 0.01	3 ± 1
3	8.6 - 11.7	30.1 - 32.2	-48.6	-	<5 ± 5	335 ± 1	0.62 ± 0.01	36 ± 5	0.01 ± 0.01	13 ± 1
4	12.1 - 15.0	28.5 - 29.9	-49.7	-	8 ± 5	334 ± 1	0.77 ± 0.01	69 ± 5	<0.1	43 ± 1
5	12.4 - 14.0	28.95- 29.75	-49.7	-	12 ± 5	333 ± 1	0.74 ± 0.01	63 ± 5	<0.2	34 ± 1
6	16.1 - 18.8	27.0 - 28.0	-51.9		<5	333 ± 1	0.81 ± 0.01	81 ± 5	0.47 ± 0.1	51 ± 1
7	19.0 - 22.1	25.9 - 26.9	-51.9	-	<5	332 ± 1	0.78 ± 0.01	80 ± 5	1.9 ± 0.1	51 ± 1
8	23.9 - 26.3	24.8 - 25.4	-52.7	-	10 ± 5	333 ± 1	0.94 ± 0.01	130 ± 5	6.6 ± 0.2	99 ± 2
9	29.6 - 36.1	22.7 - 24.0	-55.0	-	10 ± 5	332 ± 1	1.04 ± 0.01	154 ± 5	17.0 ± 0.5	130 ± 3
10	39.1 - 47.3	21.0 - 22.2	-56.8	-	9 ± 5	335 ± 1	1.16 ± 0.01	210 ± 6	45.0 ± 1.5	184 ± 4
11	51.0 - 57.5	19.75- 20.5	-57.9	-	6 ± 5	336 ± 1	1.29 ± 0.01	233 ± 7	82.0 ± 2.5	234 ± 5
12	68.0 - 76.0	18.0 - 18.7	-58.5	-	<5	336 ± 1	1.49 ± 0.02	292 ± 9	152.0 ± 5.0	311 ± 6
13	85.0 - 92.0	16.75- 17.25	-59.5	-	9 ± 5	337 ± 1	1.52 ± 0.02	290 ± 9	152.0 ± 5.0	306 ± 6
14	112.0 - 121.0	15.0 - 15.5	-61.1	-	12 ± 5	339 ± 1	1.60 ± 0.02	303 ±10	180.0 ± 6.0	329 ± 7

* US-Standard Atmosphere 1976

§ collected during ascent

Table 7 C:

Flight-No. BI/8 Date: 21. October 1985

Sample No.	Altitude [mb]	Altitude [km]*	CH_3Cl [pptv]	CH_3CCl_3 [pptv]	CCl_4 [pptv]	$C_2F_3Cl_3$ [pptv]
1§	15.0 - 21.9	26.0 - 28.5	50 ± 3	<0.4	<0.05	2.2 ± 0.15
2	7.3 - 8.1	32.6 - 33.6	33 ± 3	<0.1	<0.01	0.04 ± 0.01
3	8.6 - 11.7	30.1 - 32.2	32 ± 3	<0.1	<0.01	0.33 ± 0.02
4	12.1 - 15.0	28.5 - 29.9	40 ± 3	<0.2	<0.02	1.4 ± 0.10
5	12.4 - 14.0	28.95 - 29.75	54 ± 3	<0.3	<0.03	1.1 ± 0.08
6	16.1 - 18.8	27.0 - 28.0	46 ± 3	<0.3	<0.03	1.8 ± 0.12
7	19.0 - 22.1	25.9 - 26.9	44 ± 3	<0.4	0.17 ± 0.01	2.2 ± 0.15
8	23.9 - 26.3	24.8 - 25.4	90 ± 6	<0.7	1.6 ± 0.01	4.7 ± 0.32
9	29.6 - 36.1	22.7 - 24.0	107 ± 7	7.0 ± 0.5	5.6 ± 0.3	7.3 ± 0.50
10	39.1 - 47.3	21.0 - 22.2	150 ± 9	14.0 ± 0.9	14.0 ± 0.7	8.0 ± 0.60
11	51.0 - 57.5	19.75 - 20.5	152 ± 9	40.0 ± 2.7	32.0 ± 1.6	13.0 ± 0.90
12	68.0 - 76.0	18.0 - 18.7	208 ± 13	60.0 ± 4.0	53.0 ± 2.7	17.0 ± 1.20
13	85.0 - 92.0	16.75 - 17.25	315 ± 20	77.0 ± 5.2	54.0 ± 2.7	19.0 ± 1.30
14	112.0 - 121.0	15.0 - 15.5	433 ± 27	81.0 ± 5.4	71.0 ± 3.6	20.0 ± 1.40

* US-Standard Atmosphere 1976

§ collected during ascent

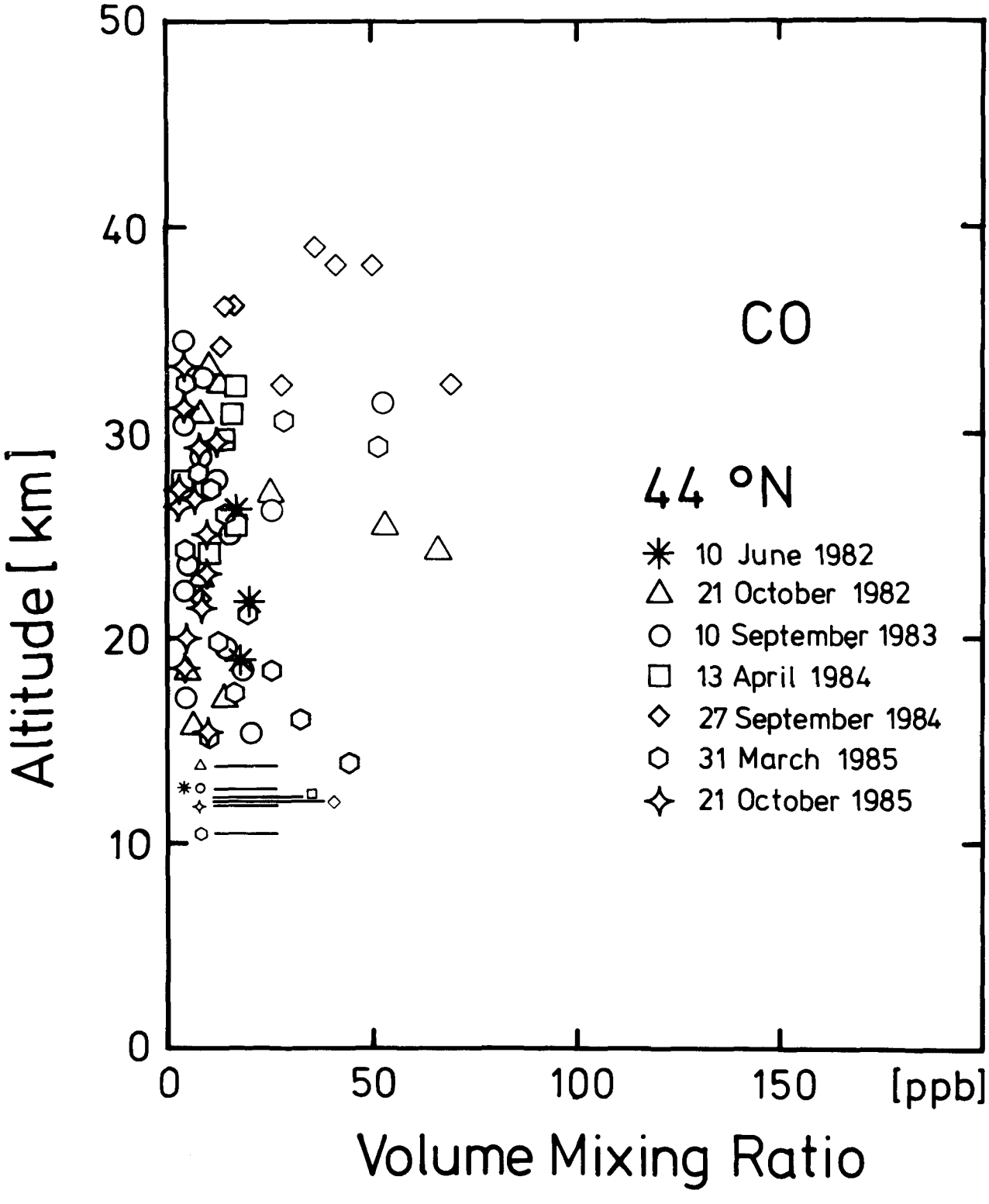

Figure 1: In situ observations of CO in the mid-latitude stratosphere 1982 - 1985. The thin horizontal bars indicate the tropopause heights for the respective flights.

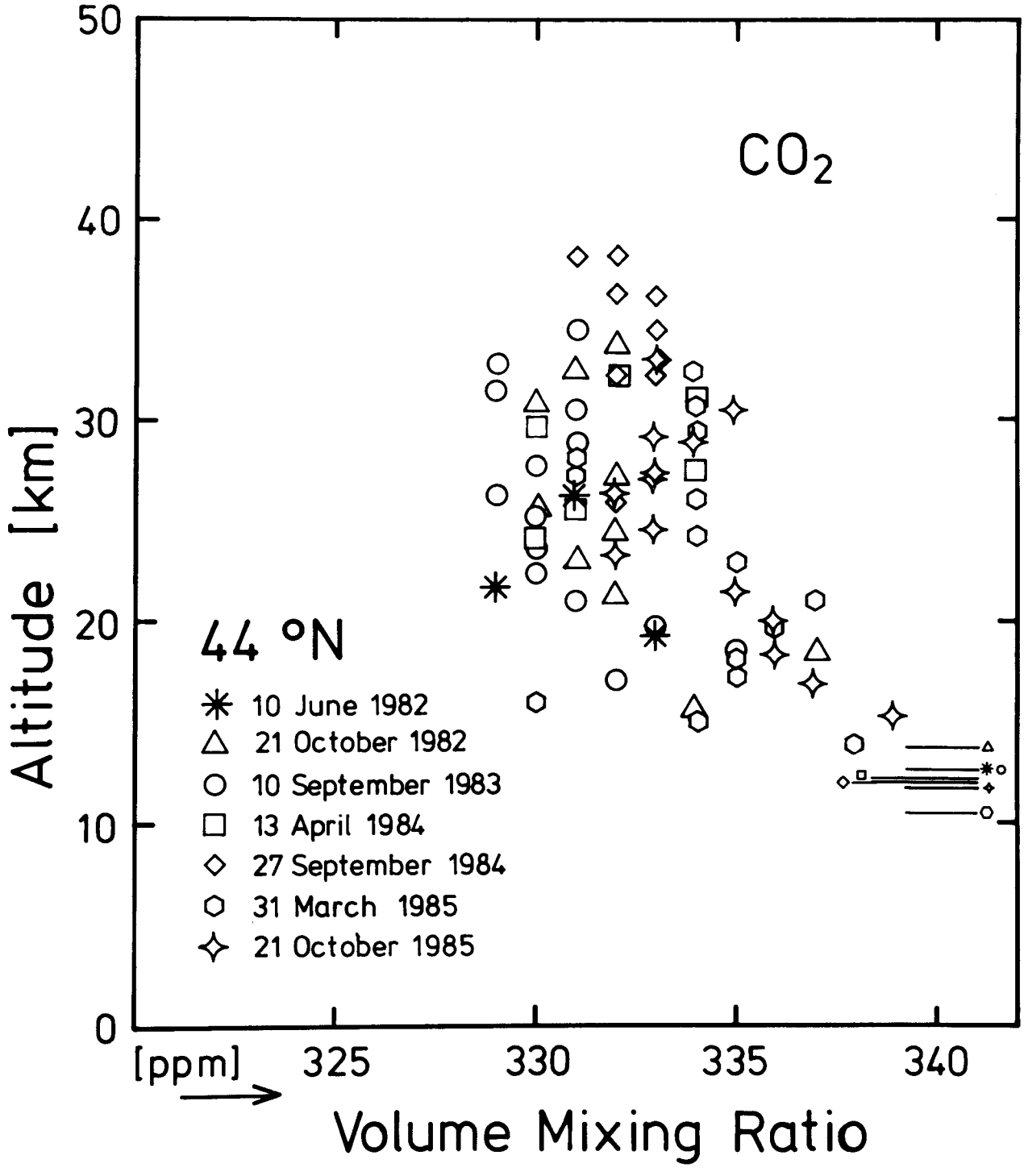

Figure 2: In situ observations of CO_2 in the mid-latitude stratosphere 1982 - 1985. The thin horizontal bars indicate the tropopause heights for the respective flights.

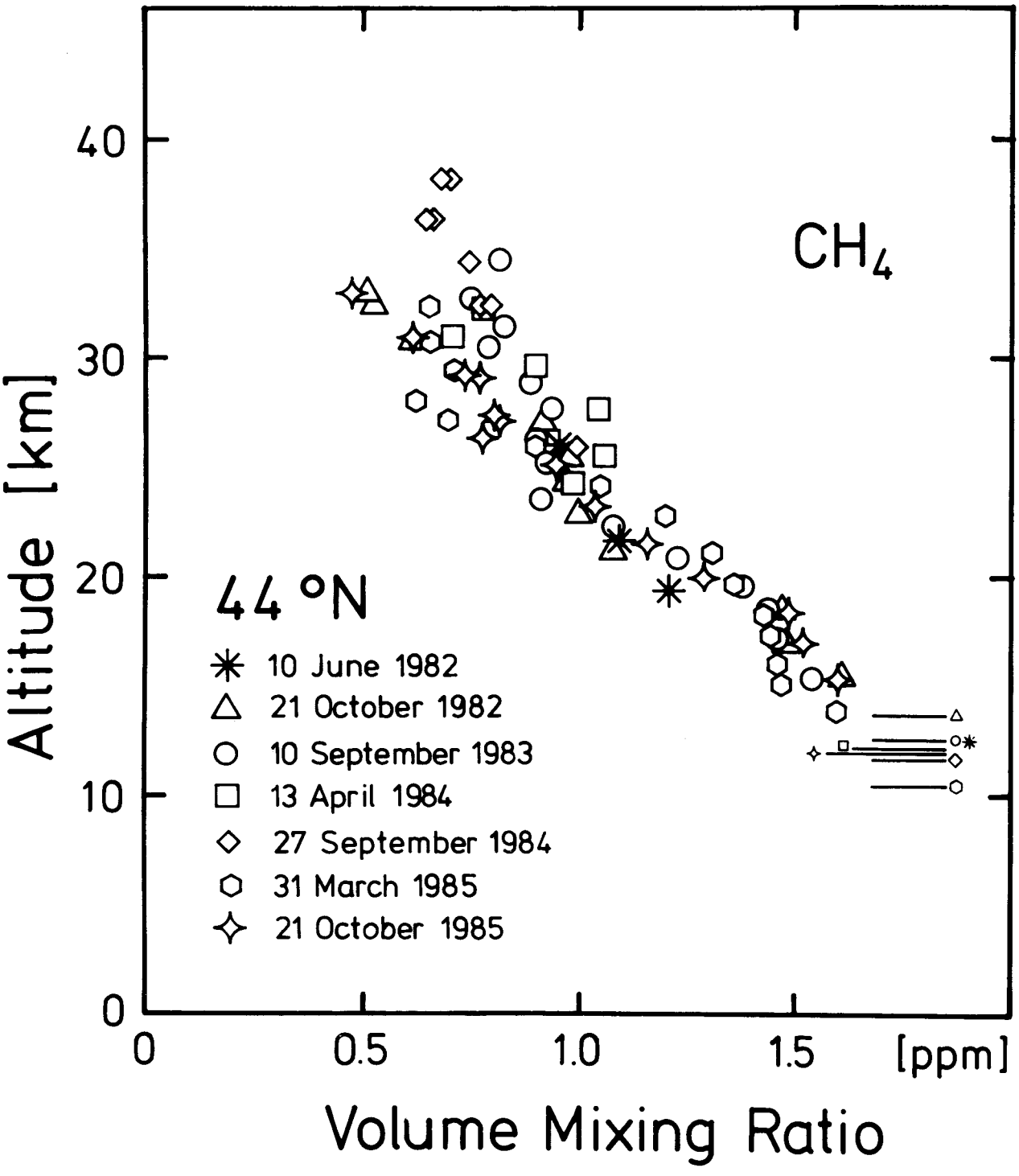

Figure 3: In situ observations of CH_4 in the mid-latitude stratosphere 1982 - 1985. The thin horizontal bars indicate the tropopause heights for the respective flights.

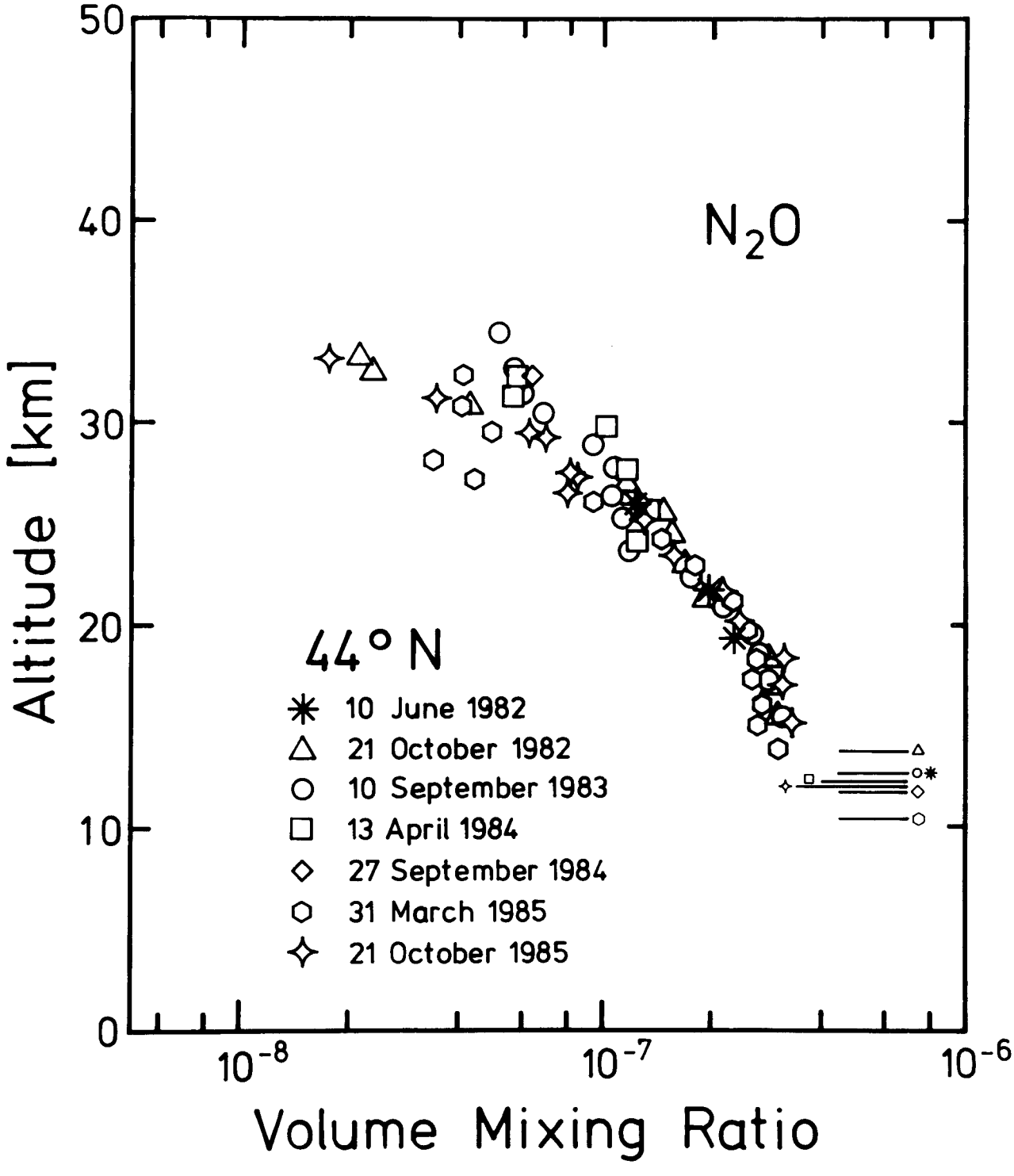

Figure 4: In situ observations of N_2O in the mid-latitude stratosphere 1982 - 1985. The thin horizontal bars indicate the tropopause heights for the respective flights.

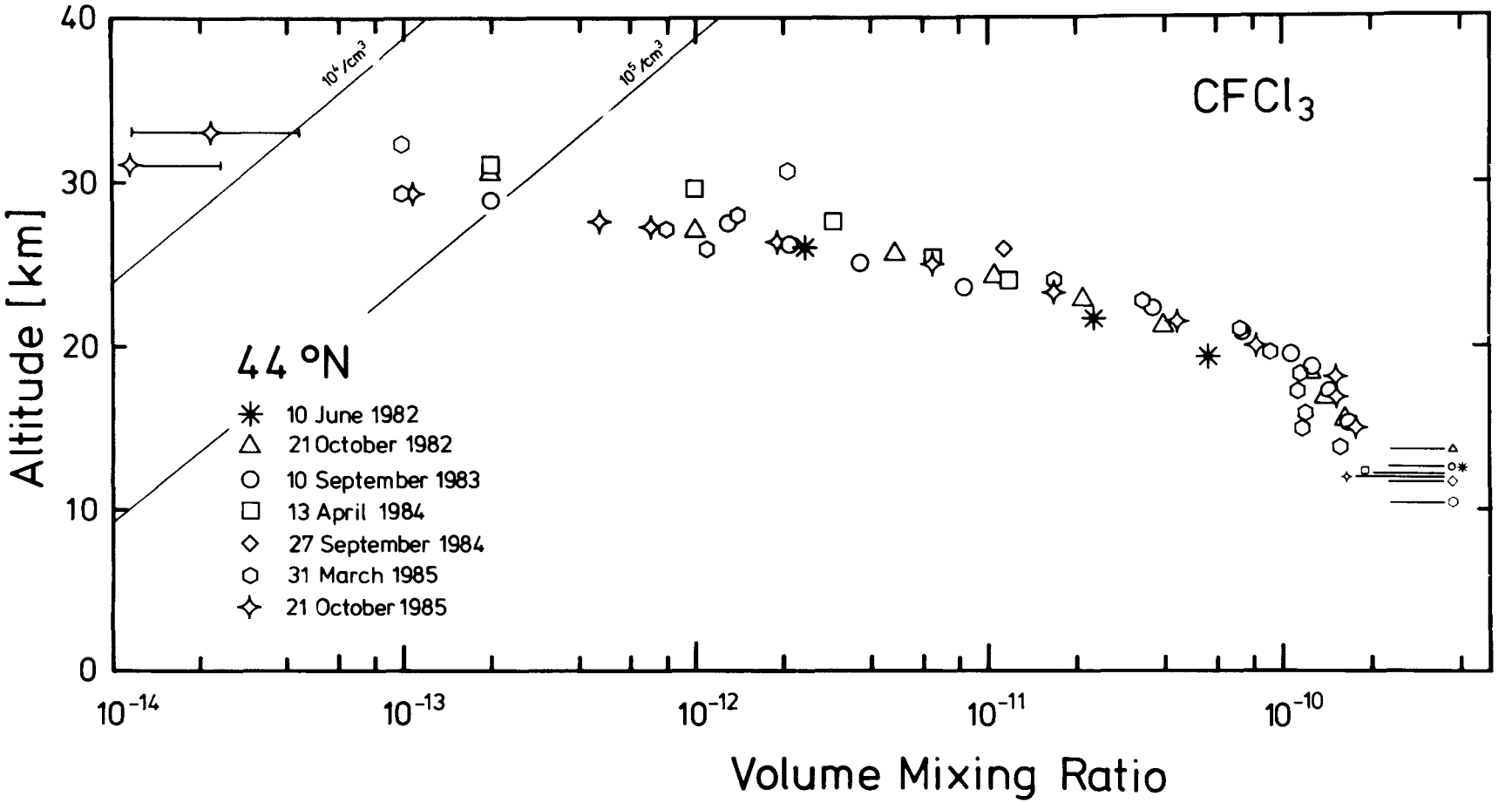

Figure 5: In situ observations of $CFCl_3$ in the mid-latitude stratosphere 1982 - 1985.
The thin horizontal bars indicate the tropopause heights for the respective flights.

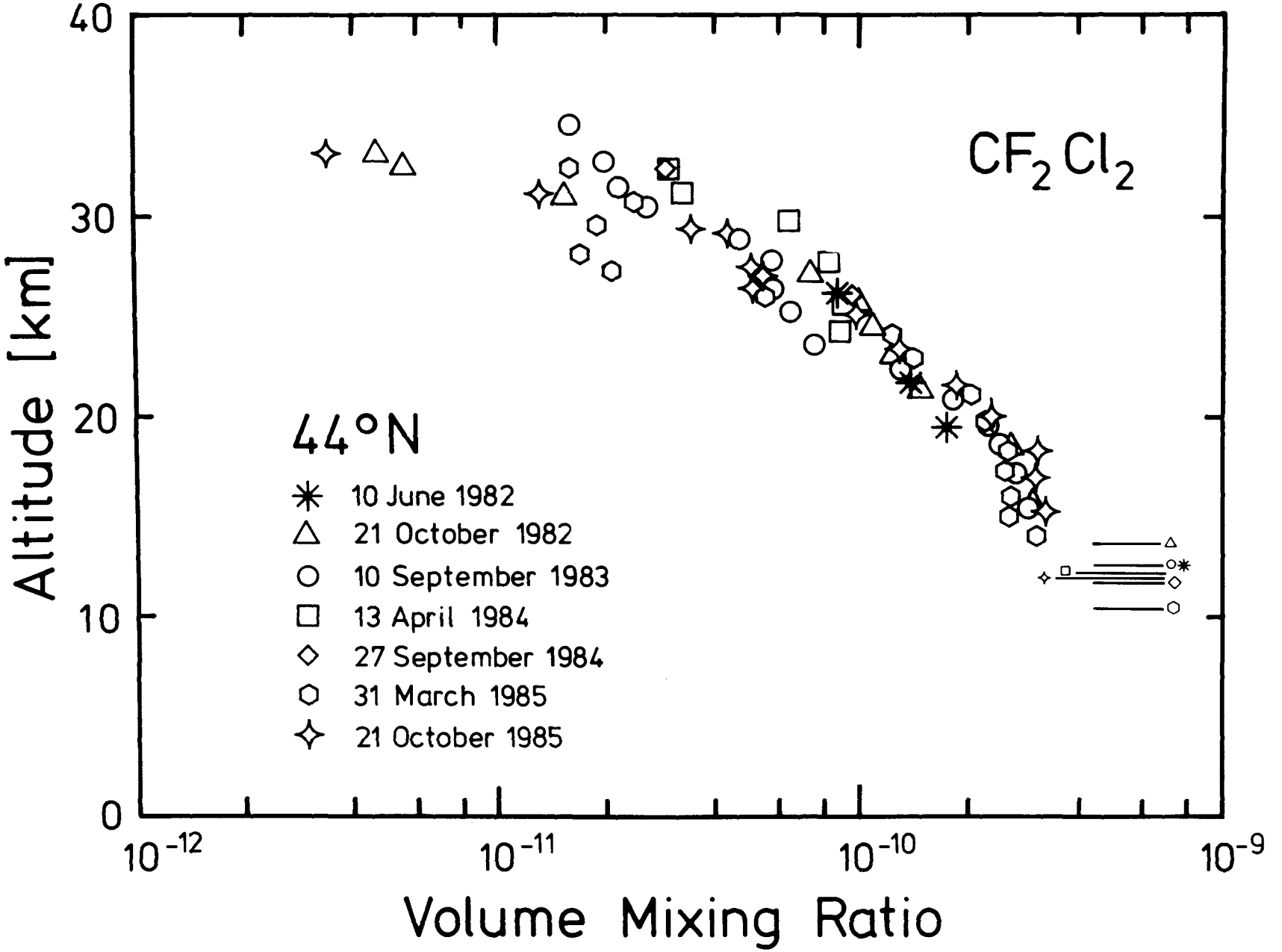

Figure 6: In situ observations of CF_2Cl_2 in the mid-latitude stratosphere 1982 - 1985. The thin horizontal bars indicate the tropopause heights for the respective flights.

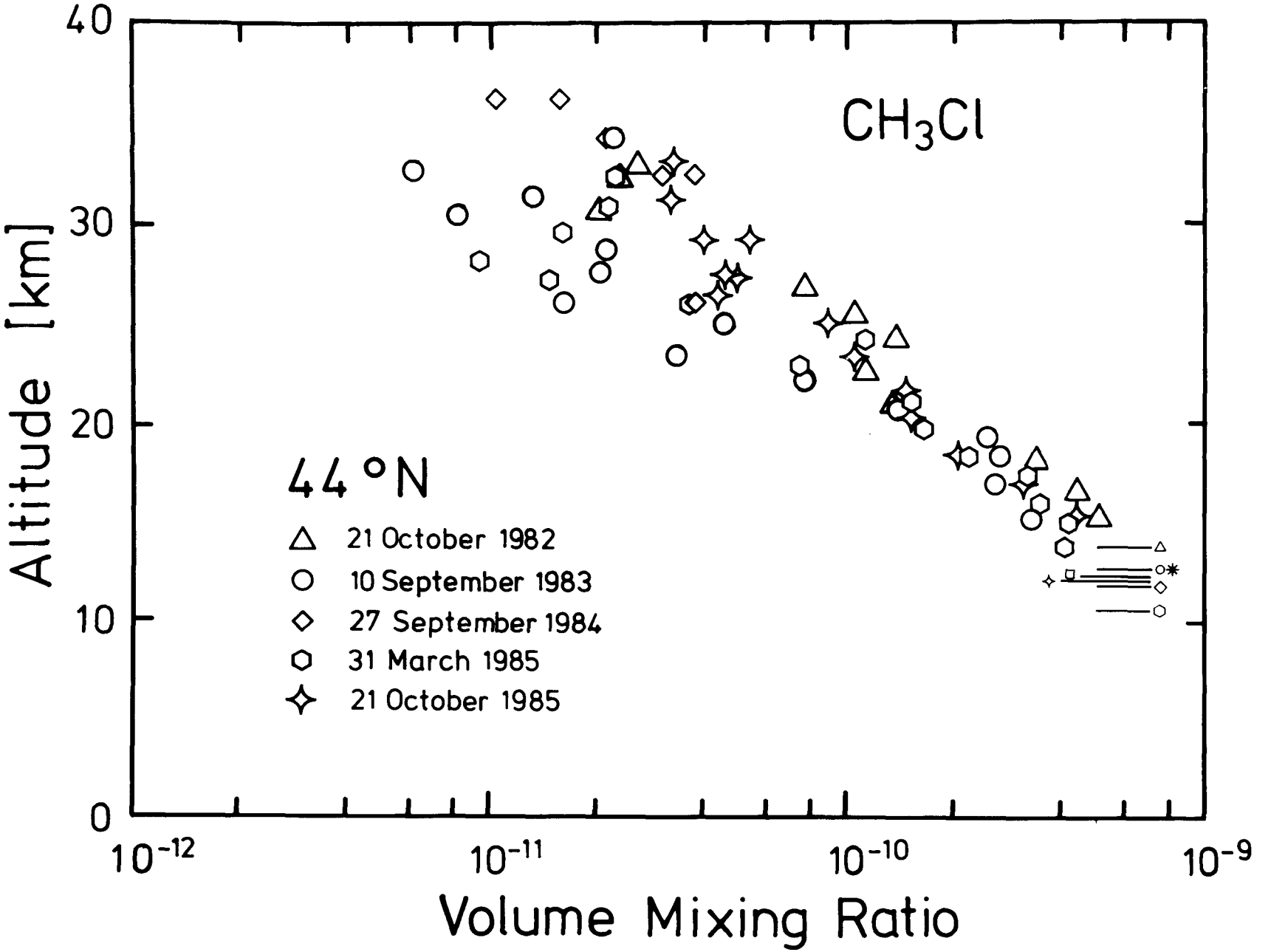

Figure 7: In situ observations of CH_3Cl in the mid-latitude stratosphere 1982 - 1985. The thin horizontal bars indicate the tropopause heights for the respective flights.

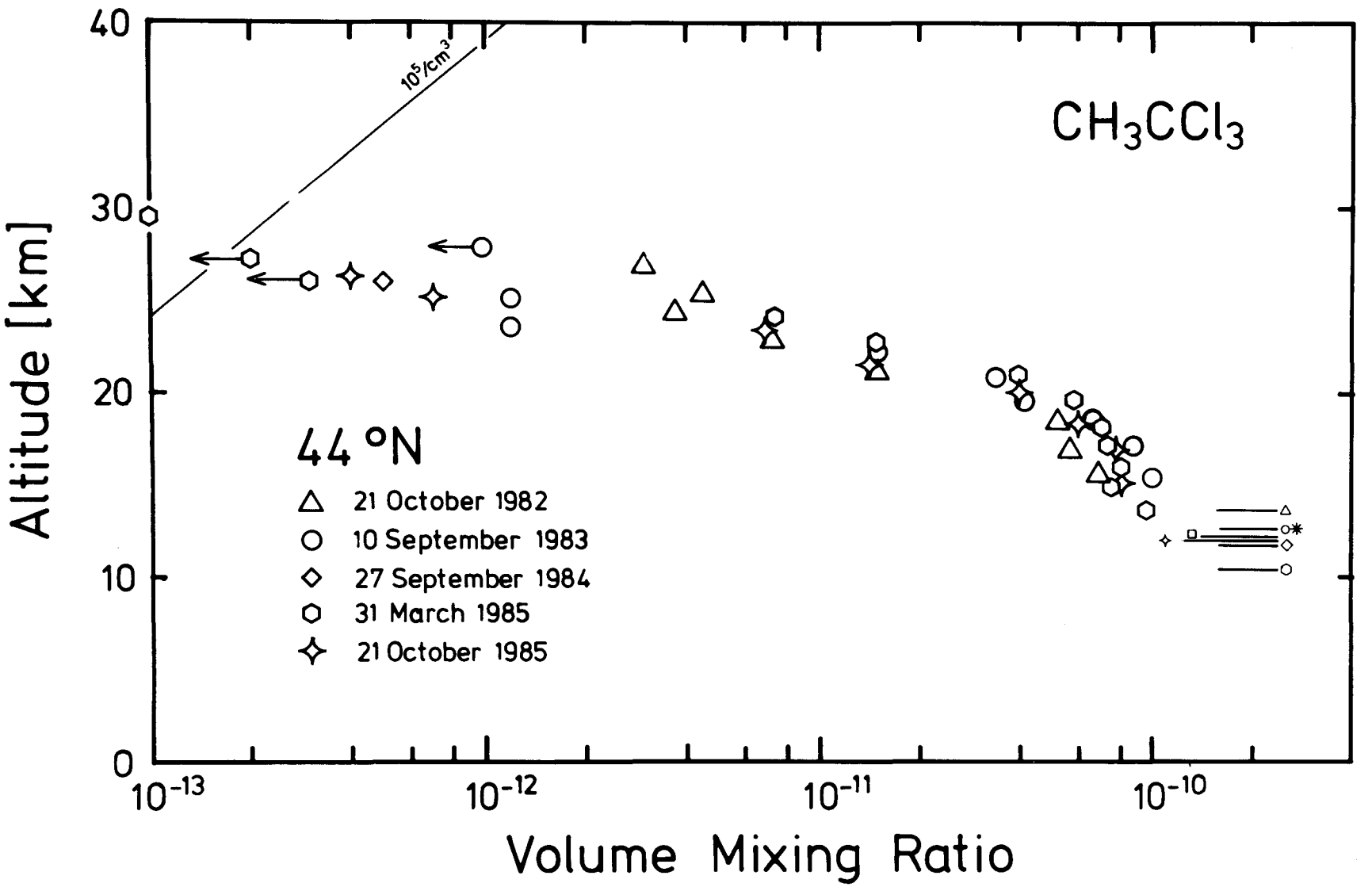

Figure 8: In situ observations of CH_3CCl_3 in the mid-latitude stratosphere 1982 - 1985.
The thin horizontal bars indicate the tropopause heights for the respective flights.

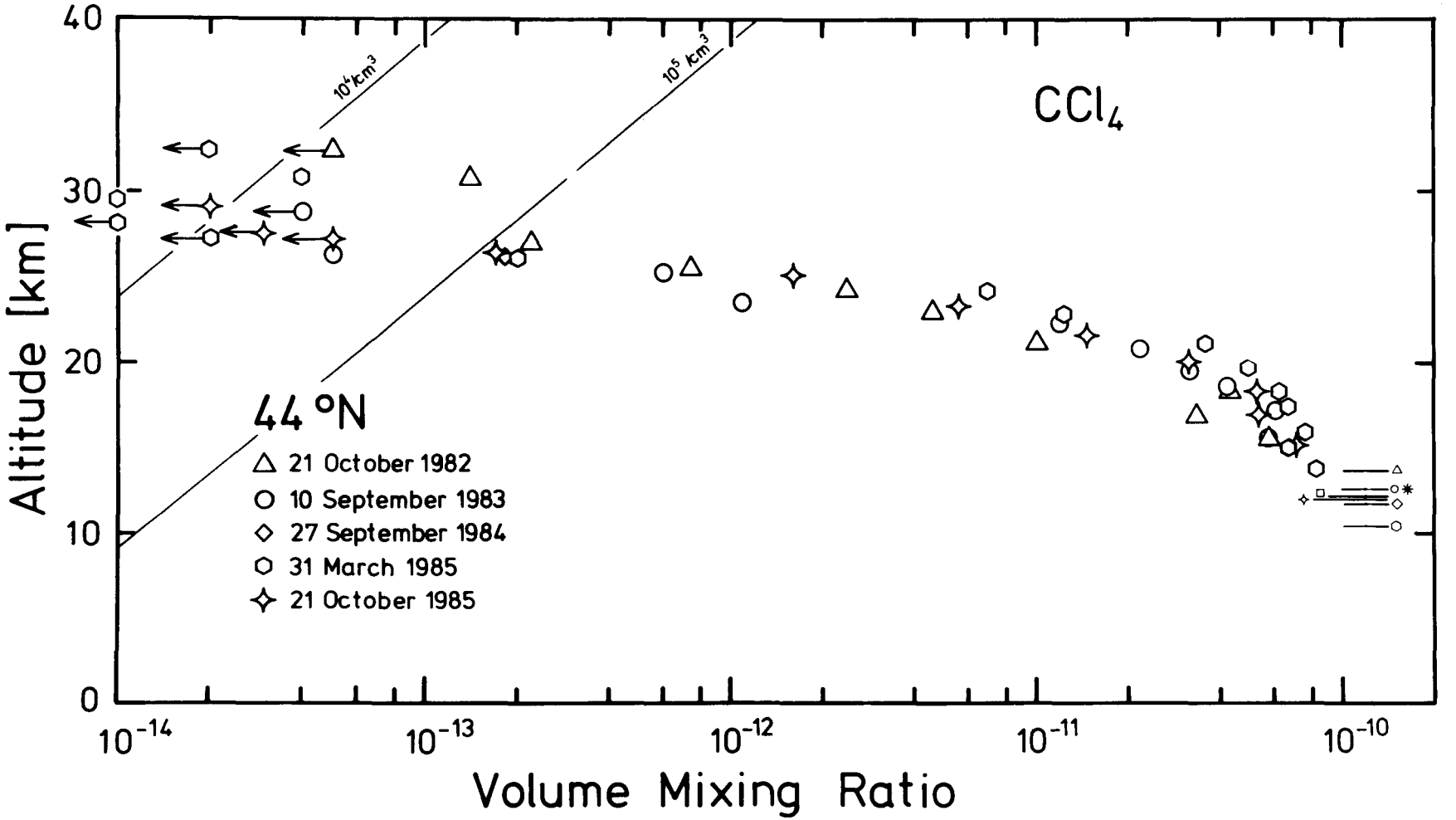

Figure 9: In situ observations of CCl_4 in the mid-latitude stratosphere 1982 - 1985.
The thin horizontal bars indicate the tropopause heights for the respective flights.

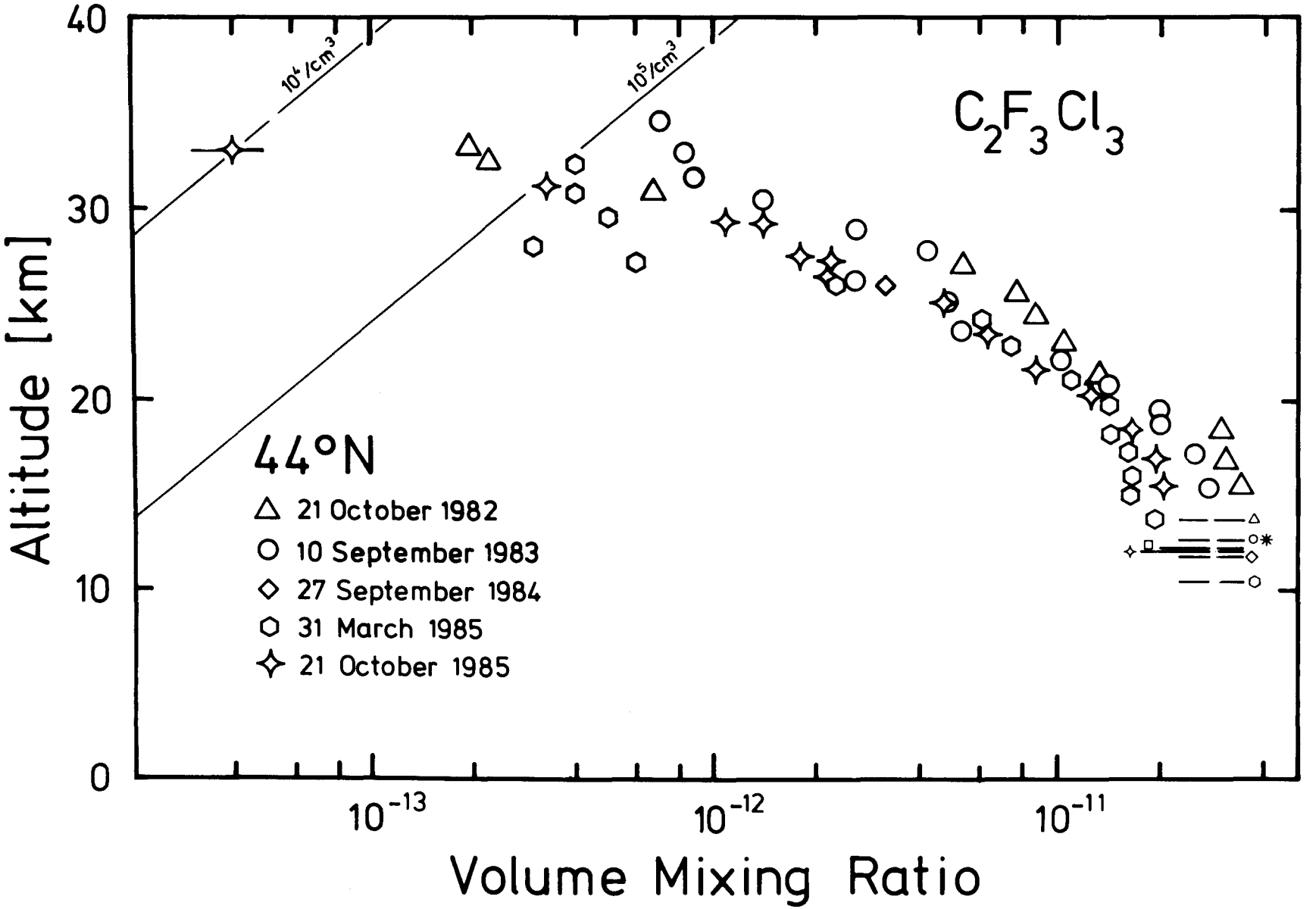

Figure 10: In situ observations of $C_2F_3Cl_3$ in the mid-latitude stratosphere 1982 - 1985. The thin horizontal bars indicate the tropopause heights for the respective flights.

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